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PESTICIDE INHALATION EXPOSURE, AIR CONCENTRATION, AND DROPLET SIZE SPECTRA FROM GREENHOUSE FOGGING

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PESTICIDE INHALATION EXPOSURE AIR CONCENTRATION, AND DROPLET SIZE SPECTRA FROM GREENHOUSE FOGGING

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ABSTRACT. *Fogger application of pesticide in greenhouses can reduce application time and applicator contact with equipment. Since fog applications use smaller and more concentrated droplets than typical wet spray applications, potential for inhalation and off-target transport hazards may be increased. This study investigated temporal characteristics of airborne pesticide after thermal fogging of a greenhouse. Droplet size spectra and concentration of airborne pesticide were measured. Volume median diameter of the suspended aerosol decreased from 14 to 2.3 µm during 1.5 h after application. Concentration of airborne pesticide decreased by 60% in the first hour after application and by 95% during 12 h after application. Implications for respiratory exposure were analytically developed from observed data.*

Hand spraying with high-pressure, high-volume systems is the most common technique of pesticide application in greenhouse culture. However, fog or aerosol applications are feasible and often-used alternatives for enclosed structures. The motivations for fog application are the potential to increase spray deposition; improve pest control efficacy; and reduce application times, rates of active ingredients, and applicator exposure. A common fogging procedure in greenhouse operations is to apply the fog in late afternoon or early evening. This practice allows cultural workers to be absent from the greenhouse during and up to 12 to 16 h after application. Also, greenhouse vents may remain closed after application since nighttime temperatures are lower. Moreover, many applications are made by positioning the fogger within the greenhouse and allowing it to run unattended until the application is complete; the applicator can remain outside the structure during application. Anecdotally, fogging is often considered a hazardous application technique since the pesticide droplets are smaller and more concentrated than in conventional "wet" spraying techniques. Such reasoning may be valid; the potential inhalation hazard may exist. Practically, the potential hazard could be mitigated through establishment of appropriate re-entry intervals and use of respiratory protection with appropriate performance criteria for workers re-entering the structure.

LITERATURE REVIEW

A comprehensive study (Brouwer *et al*., 1992) reviewed previous work (e.g., Lindquist et al., 1987) concerning airborne concentration of pesticide following low-volume and fog application in greenhouses. Brouwer observed that the number and scope of previous studies were limited and a lack of information on greenhouse environmental conditions and droplet size spectra from application equipment made comparisons between studies invalid. Brouwer experimentally determined airborne pesticide concentrations up to 10 h post-application. Volatile insecticide (dichlorvos, v.p. = 1600 mPa) and nonvolatile fungicide (thiophanate-methyl, v.p. < 0.001 mPa) were applied using a mist (nonthermal) blower with a droplet size volume median diameter of 21 µm. The decrease in airborne concentration was described by a log-linear (i.e., an exponential decay) function. Concentration of the dichlorvos decreased by 90% during the first 3 h post-application; the greenhouse was then vented and no further data recorded. Concentration of the thiophanate-methyl decreased by 99% in the 3-h post-application.

Exploratory studies of airborne pesticide concentration in greenhouses have been conducted in California by the Worker Health and Safety Branch, California Department of Pesticide Regulation (formerly California Department of Food- and Agriculture), California Environmental Protection Agency. Rech and Edmiston (1988) monitored concentration of permethrin following thermal fog application of 0.24 kg a.i./ha. Air concentrations measured 1, 2, and 13 h after application averaged 0.70, 0.60, and 0.02 mg/m3, respectively. While the temporal decay was not characterized, especially in the 0- to 1-h and 3- to 13-h post-application range, the long-term results were similar to those of Brouwer et al. who found airborne concentration had decreased by 97% at 13-h post-application.

The previous studies have not measured droplet size spectra of the airborne pesticide during and after application. The droplet size spectrum could be particularly useful in understanding fog application in greenhouses since: 1) inhalation hazard and effectiveness of mitigation procedures are strongly related to droplet size, and 2) prediction of physical pesticide movement and dissipation through deposition or venting requires droplet size spectra.

OBJECTIVES

The goal of this work was to develop a technique for using measured air concentration and droplet size data in combination with human inhalation characteristics and performance characteristics of protective devices to estimate potential respiratory exposure of greenhouse workers. The technique was applied to an example greenhouse aerosol application event. The specific, experimental objectives of the project were to: 1) characterize the temporal decay of airborne pesticide concentration following a fog application, and 2) simultaneously determine temporal changes in the droplet size spectrum of the airborne pesticide. The analytical objective was to combine pesticide concentration and droplet size information to characterize the airborne pesticide aerosol. The effectiveness of exposure mitigation techniques such as various re-entry intervals or respiratory protection could be predicted from the results.

EXPERIMENTAL PROCEDURES

The project was primarily post-application monitoring of the air within a commercial greenhouse. Permethrin was applied to an ornamental crop using commercially available thermal fogging equipment. Beginning immediately after fogging and extending for approximately 16 h, the concentration of pesticide active ingredient in the air was determined. Simultaneously, the droplet size spectrum of the applied material in the air was measured. All aspects of the study were conducted according to U.S. Environmental Protection Agency Good Laboratory Practice Standards, 40 CFR 160, amended and effective 16 October 1989 and followed sampling guidelines developed by the California Department of Food and Agriculture (Sava, 1994).

Site, Material, And Application

The test site was a 1896 m^2 (57.6 m x 32.9 m) greenhouse in Nipomo, California, used for production of potted citrus stock for ornamental use and orchard production. The structure consisted of seven bays, with each bay 8.2 m wide x 32.9 m long (fig. 1). The roof of each bay was 3.0 m high at the edges and extended to an elliptical height of 4.5 m at the center. The enclosed volume of the structure was approximately 7200 m^3. Each bay had a 1.2-m roof vent that ran along the ridge of the 32.9 m length. One side wall of each bay had a 1.2-m side vent that ran along the 8.2-m side wall. During testing, the structure was filled with 10 month old, 0.8-m-tall plants placed on 0.6-m-high benches.

The pyrethroid insecticide permethrin (CAS Registry No. 52645-53-1), provided by FMC Corporation (Philadelphia, Pa.) and formulated as Astro T&O 3.2 ECTM, was used as the test pesticide. Permethrin is a relatively nonvolatile (v.p. = 0.0013 mPa) material, registered for greenhouse use on ornamentals and often applied through fog systems. Since the study was concerned with the physical transport of pesticide droplets rather than pest control efficacy, residue breakdown or biological effects, results could be applied to other compounds and tank mixes with similar physical properties.

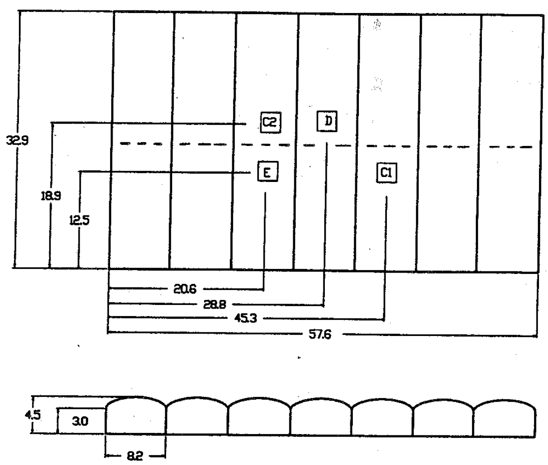

Figure 1-Plan and elevation views of test structure Droplet size probe, environmental (temperature and relative humidity) datalogger, air concentration measurement locations 1 and 2, denoted by D, E, C1, and C2, respectively. All dimensions in meters. Dotted line indicates center aisle and path of applicator.

The pesticide was applied through a thermal fogger (model K-3, Dramm International, Manitowoc, Wis.) which had been serviced and calibrated by the manufacturer prior to the experiment. The experienced applicator operated the equipment in a typical manner. The tank mix applied to the test site consisted of 190 mL of Astro T&O 3.2 EC, 1.03 1 of VK-II fogger solution (Dramm International) and 3.08 L of water. The tank mix was prepared 15 min prior to application. The application was started at 5:47 P.M. The applicator made one pass along the 2.5-m-wide center aisle of the structure and oscillated the fogger back and forth over the crop on each side of the aisle. The discharge of the fogger was directed toward the center of the 3.3-m open headspace between the top of the crop and the roof of the structure (fig. 2). The application required approximately 12.3 min.

Figure 2-Application of the material using a thermal logger. Applicator walked along center aisle while directing logger into headspace above plants.

The greenhouse vents were kept completely closed during the application and for 13 h after application. At 7:10 A.M. on the morning following the application, the roof and side vents were opened to allow natural ventilation. Vents were closed at 9:30 A.M. Temperature and relative humidity in the structure were measured by a solid state probe (model MF-100F, Rotronic Instrument Corp., Huntington, N.Y.) interfaced to a datalogger (model 21X, Campbell Scientific, Logan, Utah). Data were collected at 60-s intervals and are shown in figure 3. These data indicate that the relative humidity remained in the 80 to 90% range during the experimental period. This high level of humidity would result in relatively slow evaporation of the aqueous component of the spray mix. Since the remaining components, i.e., the pesticide and the fogging carrier had low vapor pressures, changes in droplet size causes by evaporation were expected to be relatively minor.

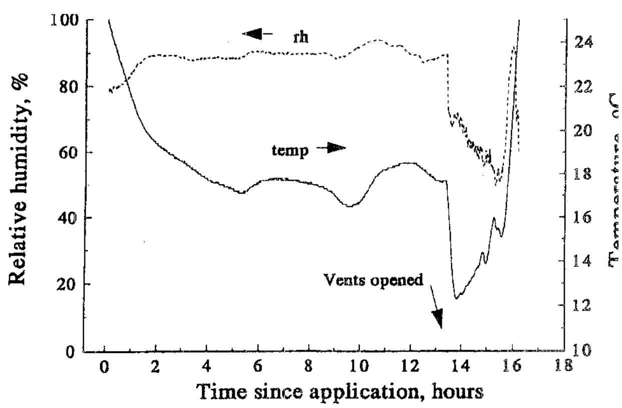

Figure 3–Temperature and relative humidity in the structure during the experiment.

Air Concentration Measurement

The concentration of permethrin in the greenhouse air was determined by sampling the air at two locations in the structure as shown in figure 1. A vacuum-type air sampler (model 114, Anderson Samplers Inc., Atlanta, Ga.) was positioned at each location and adjusted to collect 25.00 L/min. Flow rate was displayed by a rotameter indicator on each sampler. Throughout the experiment, as each sample was removed from the sampler, the rotameter indicator was inspected for any change in flow. No changes occurred and no adjustments to the samplers were made during the experiment. Flow rate for each sampler was ver led by an independent rotameter temporarily installed in the flow stream. The verification was done at the start and conclusion of the experiment.

A polypropylene filter cassette was attached to the inlet of each air sampler. Each cassette held two 47-mm-diameter glass fiber, high aerosol retention (> 99.98% with droplets down to 0.3 µm in diameter) filters (Type AE, Gelman Filters). The filters were mounted in series in the flow path to provide a primary filter and a "breakthrough" detection filter 2 mm downstream. The filters were positioned at a height of 1.5 in and oriented along a vertical plane.

Background, i.e., pretreatment, samples of 60-min duration were collected immediately before the application. The sampler air pumps were restarted immediately after the application was completed. Filter cassettes were replaced every 15 min during the first hour after application, every 30 min during the interval from 1 to 4 h after application and hourly during the interval of 5 to 16 h after application. After the cassettes were removed from the samplers, they were disassembled and the filters were removed, placed in glass jars and immediately stored on dry ice.

Permethrin was extracted from the filters by agitating the filters in 20 mL of ethyl acetate for 30 min and then injecting aliquots into a gas chromatograph (model 5880A, Hewlett Packard, Palo Alto, Calif.). An HP-1 column (10 m, 0.25 mm, 0.33 µm was used with an electron capture detector. Instrument temperatures were 220^o, 250^o, and 350^o C for the oven, injector, and detector, respectively. Retention times for cis- and trans-isomers were 7.22 and 7.56 min, respectively. Field spikes were prepared by placing 10 µL corresponding to 3.838 µg of permethrin of the formulated pesticide on filters. Average recovery from the 19 samples was 4.186 µg, or within 9% of the expected value. Detection limit for the analysis was 2 µg per sample.

Droplet Size Spectra Measurement

The size spectrum of the suspended pesticide droplets was determined using a forward-scatter laser spectrometer (model FSSP/PMS-1058B, Particle Measurement Systems, Inc., Boulder, Colo.) as described by Picot *et al*. (1985) and Sid Ahmed (1987). The system, commonly used for determination of droplet size spectra in atmospheric research and for spray clouds from aerosol nozzles, can determine droplet sizes ranging from 1.1 to 102.7 µm in diameter. The probe was located at the position shown in figure 1 with the inlet 1.2 m above the floor of the structure. The inlet was oriented horizontally and the suction fan on the probe pulled sample air into the measurement volume within the probe.

The system categorizes droplets into 16 size channels. Raw droplet counts are corrected to an actual resolution of five to six size categories using the technique of Sid Ahmed (1987) who analyzed the fundamental basis of the measurement technique. Corrected data counts from the system were used to fit a lognormal droplet size distribution. Background aerosol counts and size spectra were measured in the greenhouse prior to the fog application. Prior to the sampling, at, midpoint of the experiment and after the experiment, proper operation of die instrument was verified by passing various sizes of glass beads (Duke Scientific, Palo Alto, Calif.) through the instrument.

RESULTS
Air Concentration

Permethrin concentration in the air was calculated by dividing the recovered mass of permethrin from each primary filter by the product of the volumetric flowrate of the samplers (25.00 L/min) and the actual, observed time during which air was drawn through the filter. Only trace amounts of permethrin, near the detection limits of the analysis, were found on a few of the breakthrough filters; this confirmed that essentially all permethrin was retained on the primary filters.

Observed air concentrations during the post-application period are shown in figure 4. Data from the two sampling locations agreed closely and were used to fit a segmented model of the form:

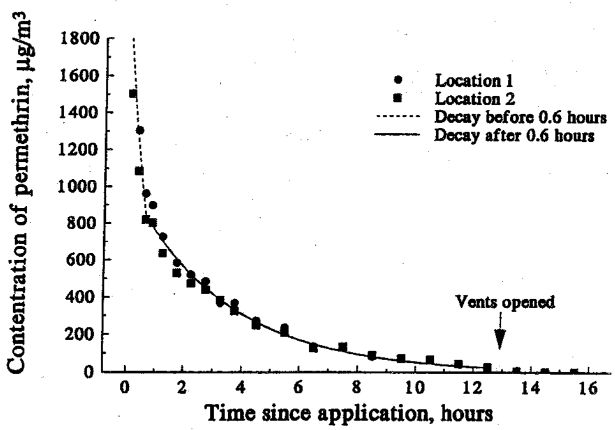

Figure 4–Concentration of permethrin in greenhouse air during experiment.

$$C(t) = C_0 \, e^{-t/\tau_0} \quad 0 < t < T$$

$$C(t) = C_1 \, e^{-t/\tau_1} \quad T<t< 13$$

where

$C(t)$ = concentration of permethrin (µg/m^3) at time t
t = time post-application (h)
T = time at which curves join (h)
C_0, C_1 = initial concentrations for each curve (µg/m^3)
τ_0, τ_1 = dissipation constants (h)

Least-squares fitting of the model yielded the parameter estimates and standard errors (in parentheses) of:

T = 0.63 (0.16)
C_0 = 2101 (102.6) τ_0 = 0.67 (0.15)
C_1 = 1050 (63.83) τ_1 = 3.26 (0.37)

Droplet Size Spectra

The calculated volume median diameter (vmd) of the suspended aerosol during the experimental period is shown in figure 5. The vmd was below 2.3 gin during the period of 1.5 to 16 h after application. The physical detection limit of the measurement system was 1.1 µm; however, the analytical technique of spectral curve fitting did not allow resolution of spectral volume median diameters below 2.3 µm. The practical implication of the data from the entire sampling period was that the droplet size of suspended pesticide in the greenhouse air decreased very quickly after application. This observation was coincidental with the observed decrease in airborne pesticide concentration during the same period. The vmd of the suspended aerosol during the first 1.5 h after application is shown in figure 6.

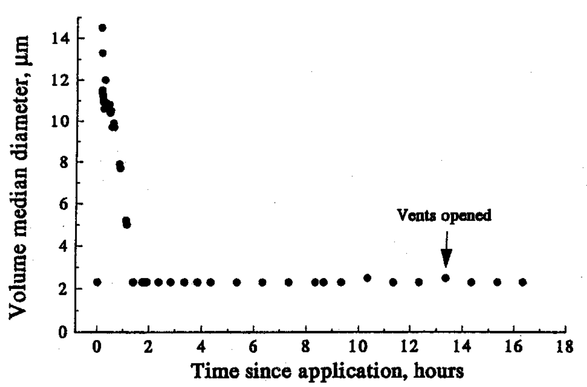

Figure 5–Observed volume median diameter of suspended pesticide aerosol in greenhouse air during entire experimental period.

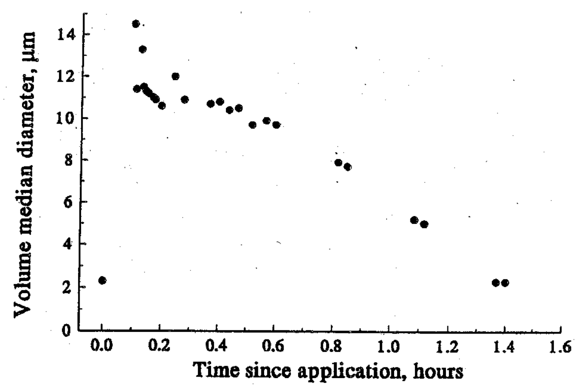

Figure 6–Observed volume median diameter of suspended pesticide aerosol in greenhouse air during initial 1.5 h after fogger application.

The droplet size spectrum of the aerosol cloud emitted from the fogger could be characterized by the spectra recorded by the measurement system as the fogger passed by during the application. Observed spectra, recorded every 30 s during the 2-min period when the fogger was passing through the area near the measurement probe, are shown in figure 7. The spectra indicate that approximately 70% of the emitted volume of spray liquid was contained in droplets which were categorized as being in the 6.9 to 15.4 µm size class. The volume median diameters of the observed spectra in figure 7 ranged from 10.9 to 14.5 gm. The log-normal distribution parameters (Hinds, 1982) of count median diameter and geometric standard deviation of the distributions ranged from 6.6 to 7.3 and 1.4 to 1.6, respectively. Droplet size spectra for the period of 0.5 to 2.5 h after application are shown in figure 8. As time after application progressed, large droplets constituted a sharply decreasing portion of the volumetric distribution.

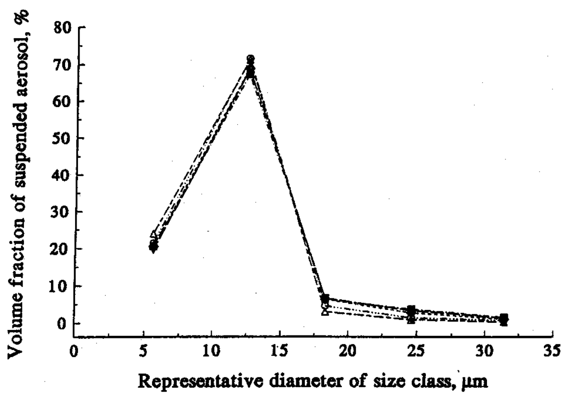

Figure 7–Droplet size spectra of emitted aerosol from thermal fogger. Samples collected at 30-s intervals during a 2-min period.

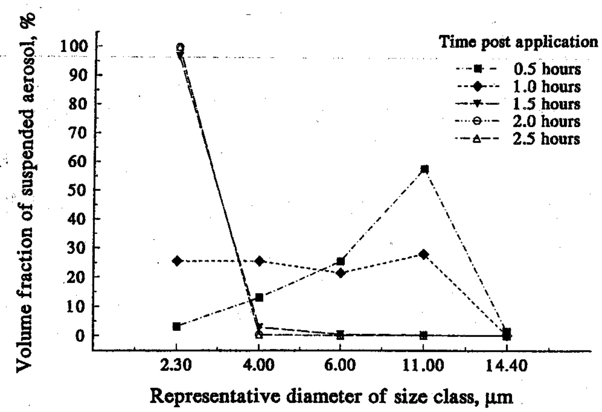

Figure 8–Temporal changes in droplet size spectra of suspended aerosol in greenhouse air during period of 0.5 to 2.5 h after application.

ANALYSIS AND DISCUSSION

Inhalation hazard and potential personnel exposure are determined by the airborne concentration of the pesticide, the droplet size spectrum of the droplets, and exposure mitigation measures which may be taken. The applicator would be exposed to the highest air concentrations of pesticide during application. Air concentrations decrease rapidly after application. Re-entry workers, if prohibited from entry into the structure during an appropriate re-entry interval, would be exposed to the lowest concentrations.

Respiratory protective devices, or respirators, are the most common exposure mitigation technique for inhalation hazards. The protection factor of a given respirator represents the reduction in contaminant concentration from the ambient atmosphere air to the air within the facepiece of the respirator. NIOSH (1987) reported Assigned Protection Factors (APF) ranging from 10 for half-mask respirators (corresponding to 90% protection) to 50 for full-face respirators (98% protection) with high-efficiency filters to 10,000 for a self-contained respirator with a full facepiece operating in a positive pressure mode (99.99% protection). While actual performance of a particular device may depend on aerodynamic diameter of the droplets or particulates in the air and the fit and maintenance of the device, the typical APFs of these devices will be used for estimates in this analysis.

Deposition of airborne particulates in the human respiratory system is dependent on the aerodynamic diameter of the inhaled material. The relationship between deposition efficiency of inhaled droplets and the droplet diameter is shown in figure 9. Essentially all droplets greater than 10 µm are deposited in the upper respiratory region of the nose and mouth. This deposition is of toxicological interest since it is ingested (Krieger, 1994). Droplets in the range of 3 to 10 µm are deposited in the tracheobronichal region with 50 to 90% of the inhaled droplets deposited. Only 20 to 30% of the inhaled droplets in the range of 0.1 to 3 µm are deposited, primarily in the alveolar region.

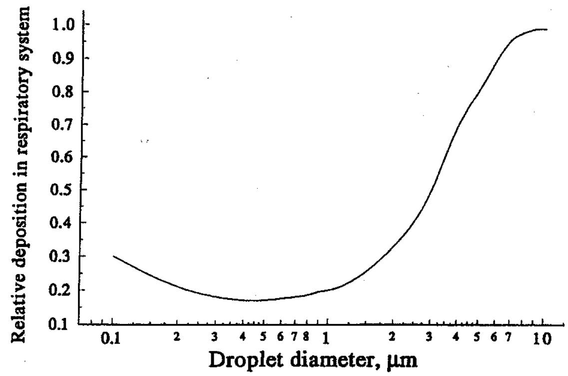

Figure 9–Effect of aerodynamic diameter on deposition efficiency of inhaled droplets in human respiratory system. Adapted from Hinds (1982).

Re-entry worker exposure is most easily reduced, by establishment of appropriate re-entry intervals or the minimum time between application and re-entry of workers into the application site. This experiment mimicked a typical mitigation strategy of making the fog application at the end of a work day, approximately 6:00 p.m., keeping the structure closed overnight and venting the structure before worker re-entry at 7:00 A.M. the next morning. Air concentration after venting was below the experimental detection limit of 1.3 µg/m^3. All detected droplets and particulates were less than 2.8 µm in diameter, indicating that only 20 to 40% of inhaled droplets would be deposited in the respiratory tract. Assuming 40% deposition of the detection limit of 1.3 µg/m^3 and a re-entry worker taking 12 breaths/min with a volume of 0.7 L/breath, the deposited mass of permethrin could be conservatively estimated at 0.26 µg/h for a worker walking into the structure. This respiration rate and volume is average for an adult female or male at rest or light work. Human ventilation rates can increase by four-fold and reach 41 L/min for males and 27 L/min for females as work effort increases and power output reaches 75 to 150 watts, respectively (CDPR, 1993; U.S. EPA, 1985).

Respiratory exposure of the applicator and workers who might enter the structure during the interval between application and venting would be dependent on the concentration of permethrin in the air, the droplet size spectra of the suspended aerosol, the APF of any respiratory protection, and the deposition efficiency of the droplets in the inhaled air. Since this study determined the former two factors and the latter two factors are well documented in the literature, potential exposure can be estimated. Estimates were developed by taking the concentration of permethrin in the greenhouse air, as estimated by the decay equations in figure 4, and determining the respiratory deposition potential. The droplet size spectra of the pesticide in the air, as shown in figures 5 through 8, and the deposition efficiency of droplets in the respiratory system, as shown in figure 9, were used to estimate the mass deposition (per unit of inhaled air) in the respiratory system. The estimated deposition could be reduced by the APF of the assumed (if any) respirator to include the effect of respiratory protection.

Results of the analysis are shown in figure 10. Potential respiratory deposition decreases rapidly after application since the air concentration and the aerodynamic diameters of the airborne droplets both decrease with time. Assuming no respiratory protection, worker breathing rate of Albumin and 0.7 L of air per breath, potential respiratory deposition would range from 9.2 jig/min immediately after application to 1.7 µg/min at 2.5 h after application to 0.07 µ/min immediately prior to venting at 13 h after application. Use of respiratory protection would decrease the potential exposures by the APF of the device. Total exposure would depend on time spent in the enclosure and worker activity.

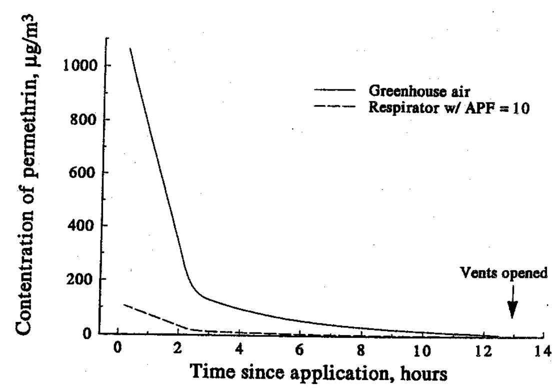

Figure 10–Airborne concentration of permethrin that could be potentially deposited in respiratory system. Calculated from air concentration, droplet size spectra of pesticide droplets, and deposition efficiency of droplets in respiratory system.

CONCLUSIONS

Concentration and droplet size spectrum of airborne pesticide were monitored after a thermal fogging application of 0.37 kg permethrin/ha. Volume median diameter of the fog application was approximately 14.5 µm. Airborne concentration of pesticide decreased rapidly (half-life - 0.46 h) in the first 0.6 h after application and more slowly (half-life - 2.26 h) during the period of 0.6 to 13 h after application. Similarly, volume median droplet size of the airborne aerosol decreased rapidly after application and was below 2.5 µm at 1.5 h after application. These observed results suggest relatively rapid settling of the suspended aerosol. Airborne concentrations were below the experimental detection limit of 1.3 µg/m3 after the greenhouse was vented at 13 h after application. Potential respiratory deposition of unprotected workers re-entering the structure after venting was estimated as less than 0.26 µg/h. Estimated potential respiratory deposition for unprotected workers entering the structure prior to venting ranged from 9.2 µg/min immediately after application to 1.7 µg/min at 2.5 h after application to 0.07 µg/min immediately prior to venting at 13 h after application. Use of respiratory protection devices would decrease these potential exposures by the NIOSH Assigned Protection Factor of the device.

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